LA-UR-21-22938
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Title: Fast-neutron multiplicity counting for nuclear safeguards and nonproliferation applications

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Intended for: Invited guest lecture at University

Issued: 2021-03-26

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Fast-neutron Multiplicity Counting for Nuclear Safeguards and Nonproliferation

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April 2nd, 2021

A little bit about me

B.S.E. Nuclear Engineering and Radiological Sciences, 2014

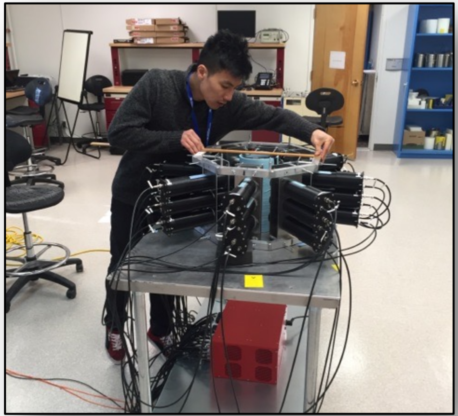
M.S.E. Nuclear Engineering and Radiological Sciences, 2015

Ph. D. Nuclear Engineering and Radiological Sciences, 2019

These title: Fast-neutron Multiplicity Counting Techniques for Nuclear Safeguards Applications

Research focus:

- Theory development for fast-neutron multiplicity counting (i.e., neutron cross-talk corrections)
- Full-system design to measure multiplicity, energy, and angular correlations in special nuclear material
- Organic scintillator detector characterization (trans-stilbene, EJ309, and organic glass)
- Multiple experimental campaigns at Idaho National Laboratory and Los Alamos National Laboratory



My LANL experience so far

Director’s Postdoctoral Fellow in the Intelligence and Space Research Division, Space Science and Applications Group (ISR-1)

Machine-learning techniques applied to optimal motion planning in multi-sensor mobile platforms

UC/LANL Entrepreneurship Postdoctoral Fellow, Feynman Center for Innovations at Los Alamos National Laboratory

Business strategy, market research, and commercialization of technical project

Motivation for Nuclear Nonproliferation and Safeguards

Nuclear technology can be used for both harmful and peaceful purposes

Castle Bravo nuclear test, 15 MT yield[1]

Enrico Fermi Nuclear Generating Station, MI U.S.A.[2]

Nuclear nonproliferation aims to **prevent the spread of nuclear weapons** and **associated technologies** through **safeguarding** of special nuclear material, ultimately to promote peaceful use

President Eisenhower delivers "Atoms for Peace" address in 1953 → international effort to develop peaceful uses

International Atomic Energy Agency (IAEA) established in 1957

The Treaty on the Nonproliferation of Nuclear Weapons (NPT) signed in 1968

The Treaty on the Nonproliferation of Nuclear Weapons (NPT)

The Treaty on the Nonproliferation of Nuclear Weapons

"Prevent the **spread of nuclear weapons and weapons technology,** to promote cooperation in the **peaceful uses of nuclear energy,** and to further the goal of achieving **nuclear disarmament and general and complete disarmament**"

First Pillar: Nonproliferation

Second Pillar: Disarmament

Third Pillar: Peaceful use

Nuclear Weapon States (NWS)

United States, Russia, China, France, and United Kingdom

Nonnuclear Weapon States (NNWS)

All other signatories

NWS Agreement

("Negotiate in good faith")

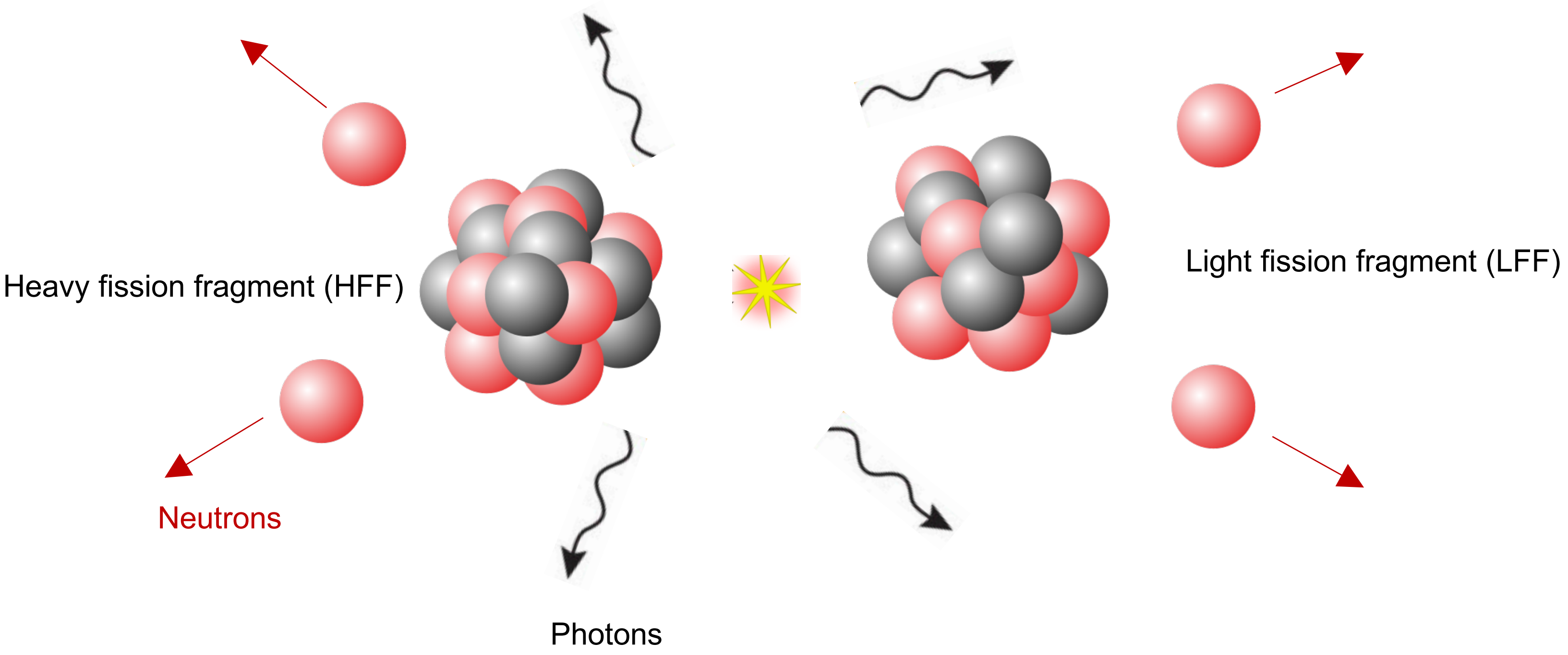
1. Effective measures relating to cessation of the nuclear arms race
2. Move towards nuclear disarmament → complete disarmament

NNWS Agreement

1. Cease efforts to acquire or access control over nuclear weapons, nuclear explosive devices, or associated technology from any NWS
2. Pledge to accept inspections from the International Atomic Energy Agency (IAEA) to verify peaceful use of nuclear technology

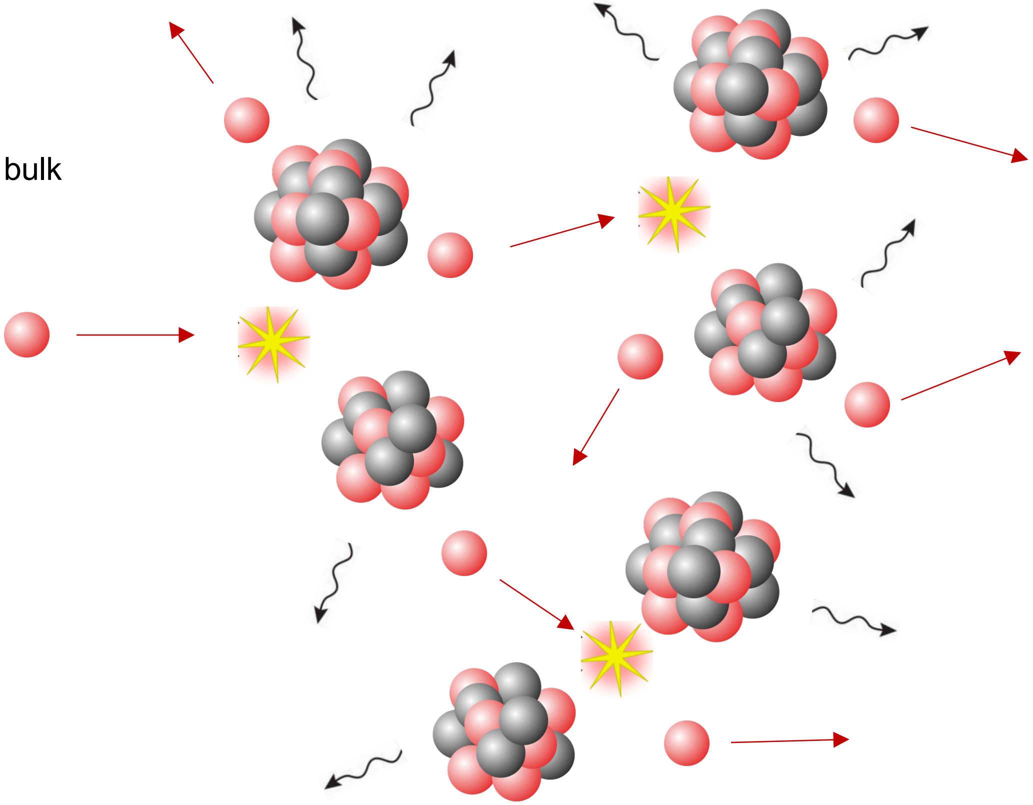
Physics of Fission

There are several characteristic signatures that arise from the fission process



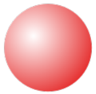
Physics of Fission

Chain reactions in bulk materials



Information rich! But difficult to detect…

Neutron and Photon Correlations



Neutron correlations

Number (i.e., multiplicity) distribution

Multiple neutrons emitted simultaneously

Energy distribution

Shared energy dissipation

Spatial/angular distribution

Momentum bias from HFF and LFF

How much is there?

Photon correlations

Number (i.e., multiplicity) distribution

Multiple photons emitted simultaneously

Energy distribution

Shared energy dissipation + characteristic spectra

What exactly is in there?

Characterizing Special Nuclear Material (SNM) by Neutron Detection

Materials Control and Accountability (MCA)

Can we verify the declared *amount* of SNM?

Can we detect *diversion* of SNM?

Nuclear Disarmament

Can we ensure nuclear dismantlement while *protecting sensitive information*?

Neutron multiplicity counting (NMC) system and techniques

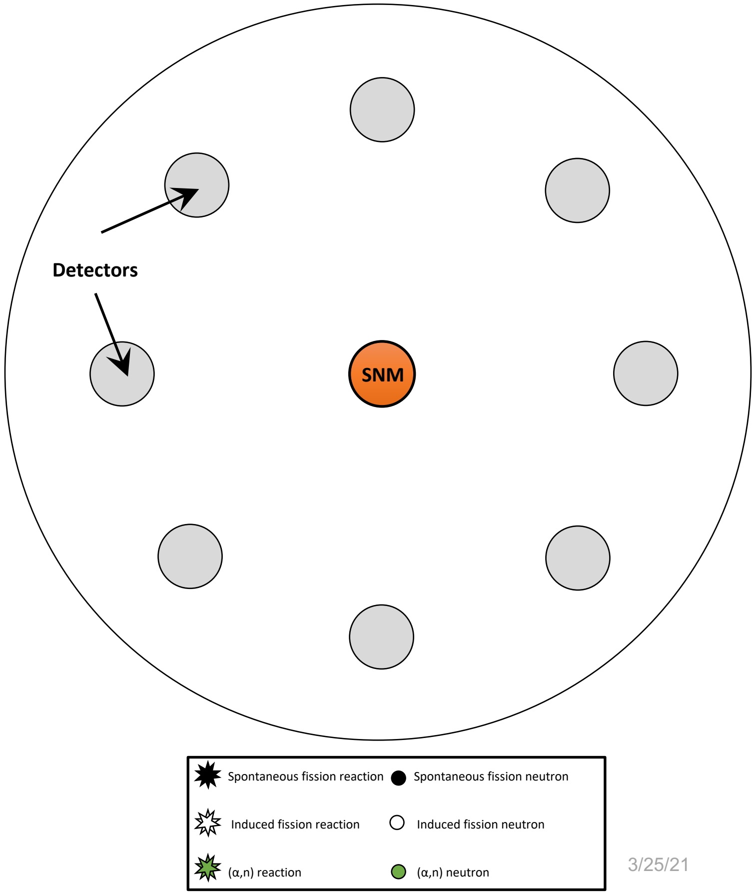
SNM emits

- Multiplets of fission neutrons correlated in time to the initiating event (**spontaneous** and/or **induced**)
- Single uncorrelated neutrons from nonfission reactions (i.e., **(α,n) reactions**)

Neutron multiplicity counting (NMC) techniques analyze the measured time-correlated neutron multiplicities to estimate physical properties of the SNM

Physical properties related to the detected **number** (i.e., multiplicity) of neutrons

1. **Effective fissile mass (F)**
   → spontaneous fission neutrons
2. **Multiplication (M_L)**
   → induced fission neutrons
3. **Contribution from nonfission neutrons (α-ratio)**
   → (α,n) neutrons

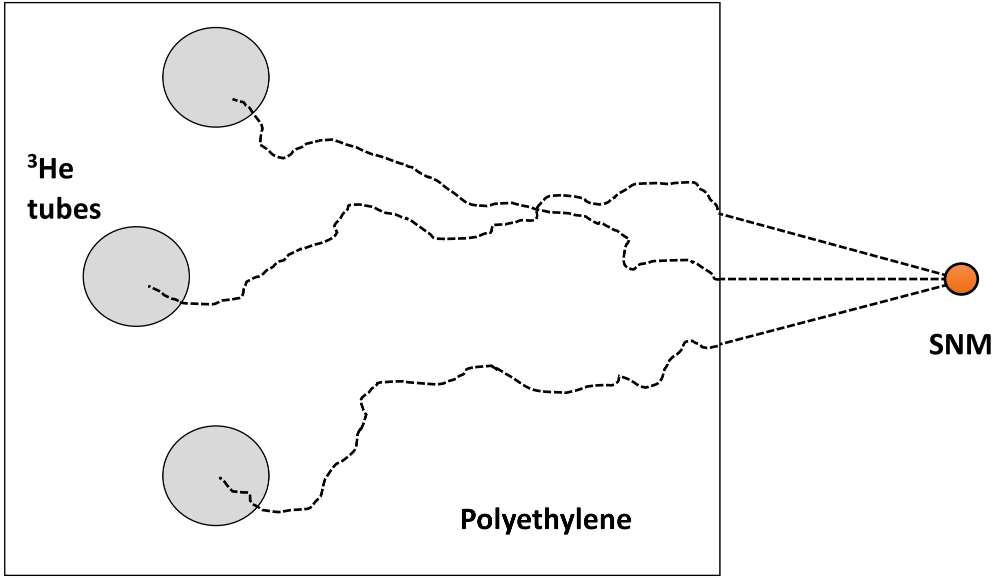


Currently deployed systems: ^{3}He-based systems

^{3}He tubes in polyethylene

Uranium Neutron Collar (UNCL)

JCC-51 Active Well Coincidence Counter



Benefits:

1. High efficiency
2. Mechanically robust
3. Performance stability/reliability

Drawbacks:

1. Long die-away times
2. Poor energy sensitivity
3. No spatial sensitivity

Current NMC method

Observable Signatures in
Multiplicity
Energy
Angle

Average correlations + event-by-event correlations

SNM Physical Properties
Mass
Multiplication
Nonfission-neutron contribution

Can we leverage the energy and angular/spatial signatures to supplement current methods for NMC?

Fast-Neutron Multiplicity Counting (FNMC)

Organic Scintillators for Fast-Neutron Detection

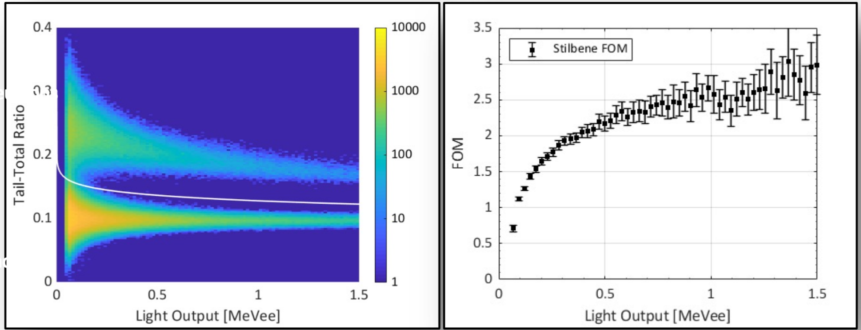
Scatter-based recoil detectors

→ Neutrons elastic scatter on protons

→ Photons Compton scatter on electrons

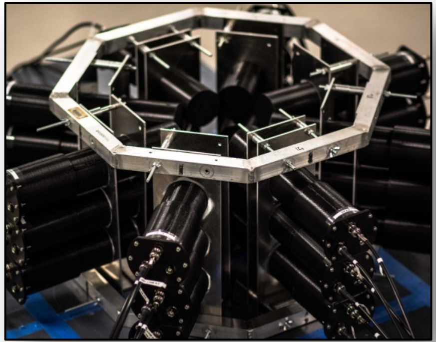
Sensitive to fast unmoderated fission neutrons

Retains a portion of initial neutron energy

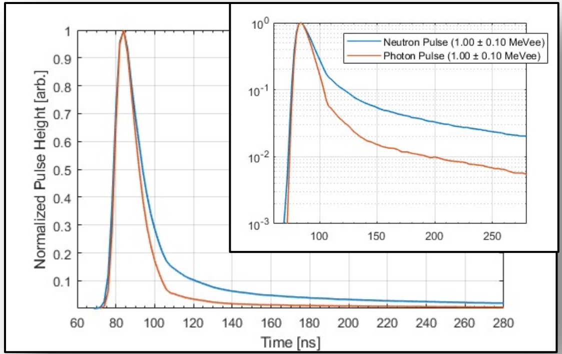


Excellent pulse-shape discrimination capability
(10^{-6} gamma-ray misclassification rate per incident detection)

trans-stilbene detector assembly



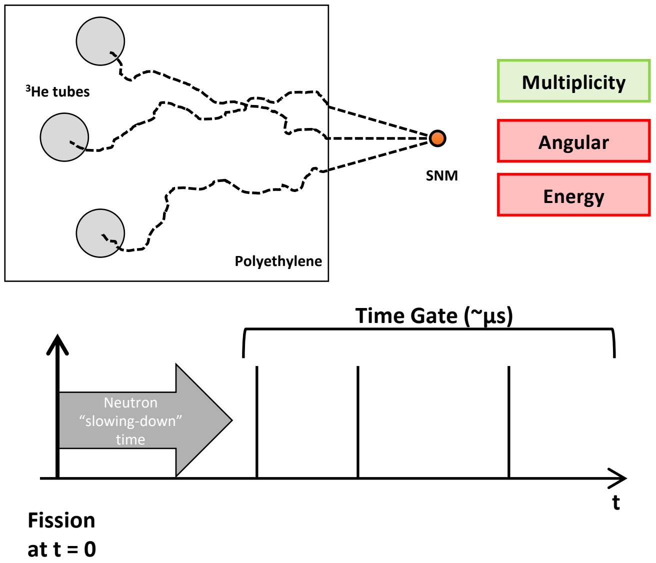
Fast-neutron multiplicity counter (FNMC)



Fast detector response (pulse width approx. 10 ns)

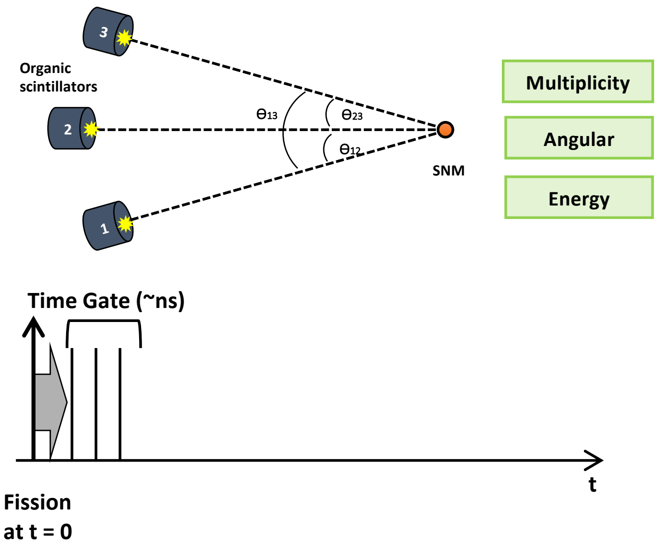
Capture-based NMC systems

Requires moderating material



Scatter-based FNMC systems

No moderating material



Benefits:

1. Shorter time gates (on the order of 10^{-3} reduction) for improved precision
2. Initial direction of emitted fission neutron retained
3. Portion of initial energy of fission neutron retained

Drawbacks:

1. Neutron cross-talk effects
2. Performance stability
3. Mechanical robustness and longevity
4. Photon sensitivity, may require shielding

MCA:

Can we verify the declared amount of SNM?

Analytic estimation of fissile mass

The point kinetics equations

$^{240}Pu_{eff}$ fissile mass can be analytically estimated from measured neutron singles (***S***), doubles (***D***), and triples (***T***) count rates using the point kinetics equations

Multiplicity count rates related to ^{240}Pu spontaneous fission rate (***F***), leakage multiplication (***M_L***), and ratio of neutrons from (α,n) reactions to those from spontaneous fission reactions (***α***) through point kinetics equations

Known quantities:	Measured quantities:	Estimated quantities:
$\nu_{sf1,sf2,sf3}$, $\nu_{if1,if2,if3}$, *system efficiency* $\boldsymbol{\varepsilon}$, *and system parameters*	*S, D, and T*	*M_L, F, and α*

$$S = \boldsymbol{f}(F,\ M_L,\ \alpha,\ \boldsymbol{\varepsilon},\ \boldsymbol{known\ quantities})$$

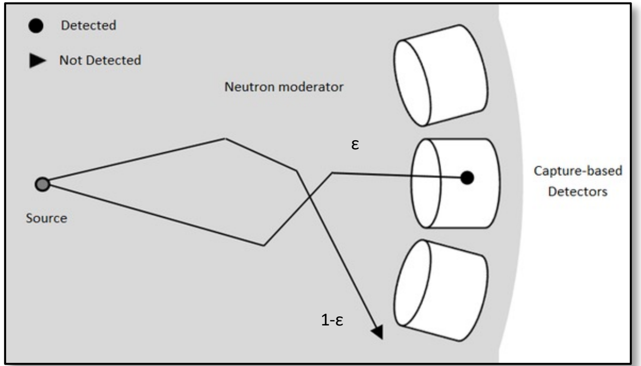
$$D = \boldsymbol{f}(F,\ M_L,\ \alpha,\ \boldsymbol{\varepsilon},\ \boldsymbol{known\ quantities})$$

$$T = \boldsymbol{f}(F,\ M_L,\ \alpha,\ \boldsymbol{\varepsilon},\ \boldsymbol{known\ quantities})$$

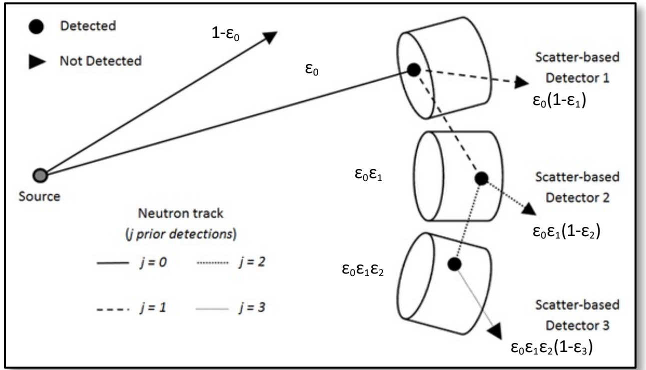
Solve for $\boldsymbol{F}$

$$\boxed{m_{^{240}Pu_{eff}} = \frac{\boldsymbol{F}}{473\,\frac{fps}{g}}}$$

Organic Scintillators for Fast-Neutron Detection



Possible detection for a single neutron in a **capture-based** system. A single neutron can only register a single count.



Possible detection for a single neutron in a **scatter-based** system. A single neutron can register multiple counts.

$$\left.\begin{aligned} S &= f(F, M_L, \alpha, \varepsilon_0, \varepsilon_1, .. \varepsilon_N, known\ quantities) \\ D &= f(F, M_L, \alpha, \varepsilon_0, \varepsilon_1, .. \varepsilon_N, known\ quantities) \\ T &= f(F, M_L, \alpha, \varepsilon_0, \varepsilon_1, .. \varepsilon_N, known\ quantities) \end{aligned}\right\}$$

Solve for F

$$m_{^{240}Pu_{eff}} = \frac{F}{473\frac{fps}{g}}$$

Passive NDA measurements of Pu-metal plates
Experiment details

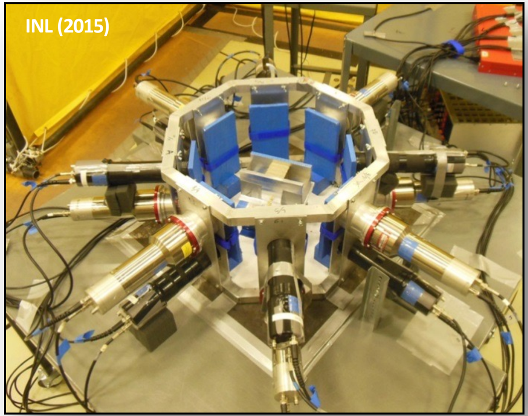
FNMC system consisted of

- 8 - 3”x3” EJ-309 liquid organic scintillators (0.52 MeV neutron-equivalent energy threshold)
- 8 - 2”x2” stilbene crystals (0.36 MeV neutron-equivalent energy threshold)

Analyzed data from two configurations of Pu-metal plates

- $^{240}Pu_{eff}$ mass of 4.72 and 14.16 g
- 0.5” lead shielding

Goal: estimate fissile mass using neutron singles, doubles, and triples rate with neutron cross-talk corrections.



Passive measurements with 8 EJ-309 and 8 stilbene

Measured neutron singles, doubles, and triples count rates

Number of plates	$^{240}Pu_{eff}$ mass [g]	Singles rate [cps]	Doubles rate [cps]	Triples rate [cps]
1	4.72	428.5 ± 0.5	11.63 ± 0.08	0.373 ± 0.014
3	14.16	1349.3 ± 1.2	46.60 ± 0.16	2.057 ± 0.034

Analytic estimation of fissile mass

With and without neutron cross-talk corrections

$$F_{CNMC} = \frac{\frac{2D}{\varepsilon_0 f^2} - \frac{M_L(M_L-1)v_{if,2}S}{v_{if,1}-1}}{\varepsilon_0 M_L^2 v_{sf,2}}$$

No. of PANN plates	$^{240}Pu_{eff}$ mass [g]	Estimated $^{240}Pu_{eff}$ mass [g]	Percent difference from true mass
1	4.72	5.35 ± 0.18	**13.35 ± 3.81 %**
3	14.16	16.97 ± 0.32	**19.84 ± 2.26 %**

$$F_{FNMC} = \frac{\frac{2D}{(\varepsilon_0+\varepsilon_0\varepsilon_1)f} - S\left[\frac{M_L(M_L-1)v_{if,2}}{v_{if,1}-1} + \frac{2\varepsilon_0\varepsilon_1}{(\varepsilon_0+\varepsilon_0\varepsilon_1)^2}\right]}{(\varepsilon_0+\varepsilon_0\varepsilon_1)M_L^2 v_{sf,2}}$$

No. of PANN plates	$^{240}Pu_{eff}$ mass [g]	Estimated $^{240}Pu_{eff}$ mass [g]	Percent difference from true mass
1	4.72	4.51 ± 0.21	**-3.81 ± 4.45 %**
3	14.16	14.55 ± 0.36	**2.75 ± 2.54 %**

FNMC can accurately estimate fissile mass with neutron cross-talk corrections

MCA:

Can we detect diversion of SNM?

Active NDA measurements of UO_2 assemblies
Experiment details

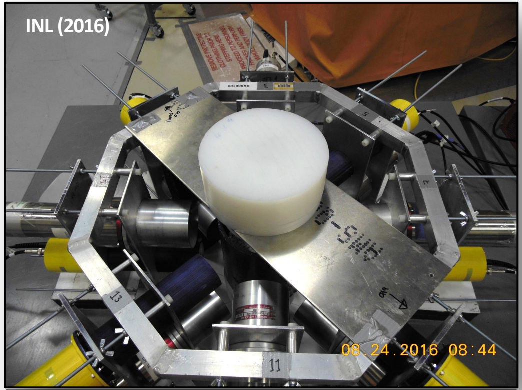
FNMC system consisted of

- 8 - 3"x3" EJ-309 liquid organic scintillators
- 8 - 2"x2" stilbene crystals
- 2 - AmLi interrogation neutron sources (5 x 10^4 n/s)
- 0.25" lead shielding

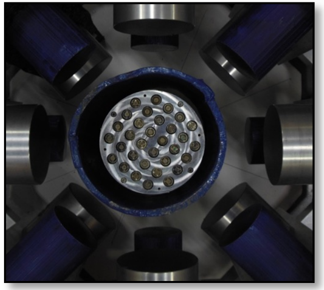
UO_2 pins arranged in fuel-like geometry, total of 32 pins

- Total uranium mass = 79.52 g
- 16.37 wt% enriched ^{235}U
- 6" length x 0.374" diameter

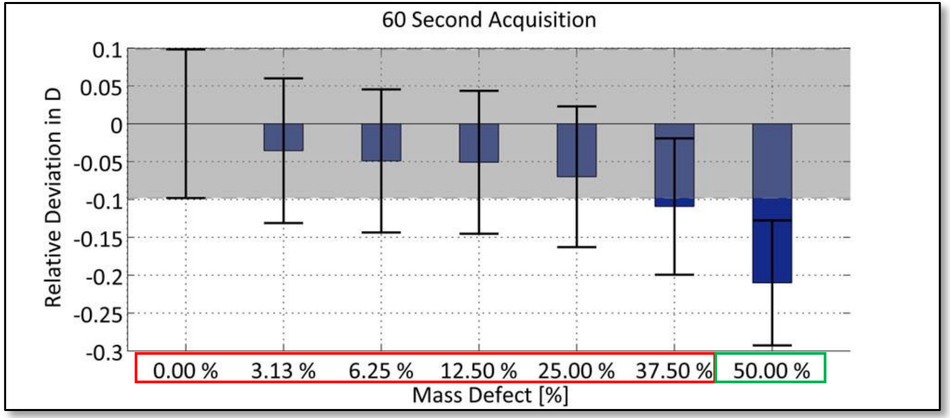
Goal: characterize FNMC sensitivity of detecting mass diversion using neutron doubles rate

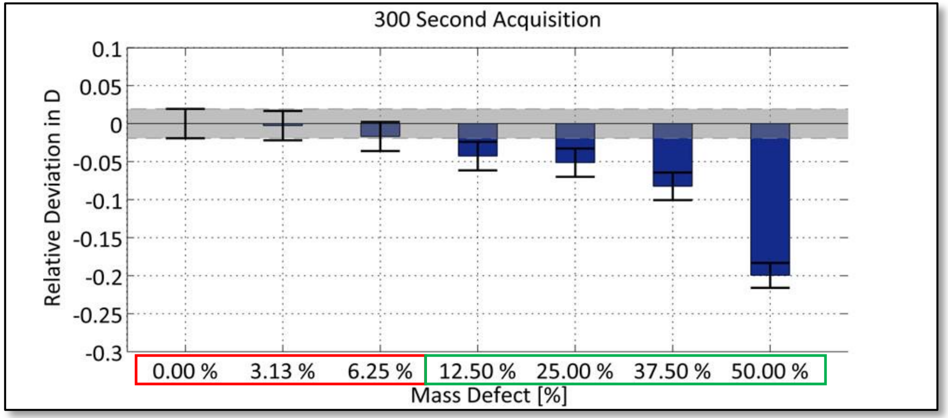


Active measurements with 8 EJ-309 and 8 stilbene


Full UO_2 assembly placed inside the FNMC system

Material Diversion Sensitivity



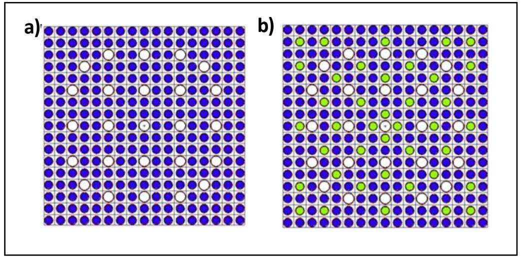


60-second assay:
→ Diversion of **50.00 %** mass detected (1272.32 g removal)

300-second assay:
→ Diversion of **12.50 %** mass detected (318.08 g removed)

Comparison to IAEA System

Benchmarked MCNP simulations of the IAEA Uranium Neutron Collar (UNCL) used to compare performance to the FNMC system



Simulated 17 x 17 fresh fuel assemblies showing the full (a) and the 15% diverted (b) configurations. The blue indicates LEU fuel rods, and the green indicates the DU replaced rods.

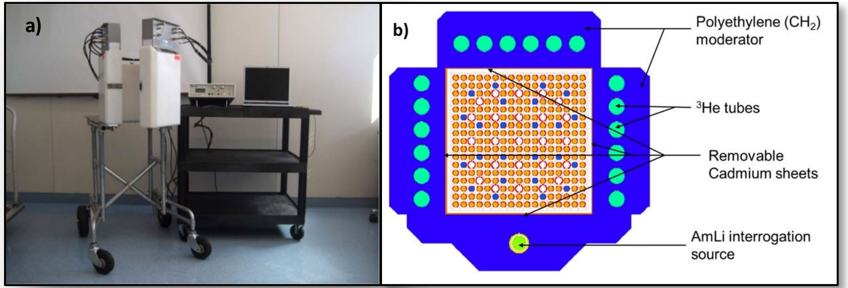
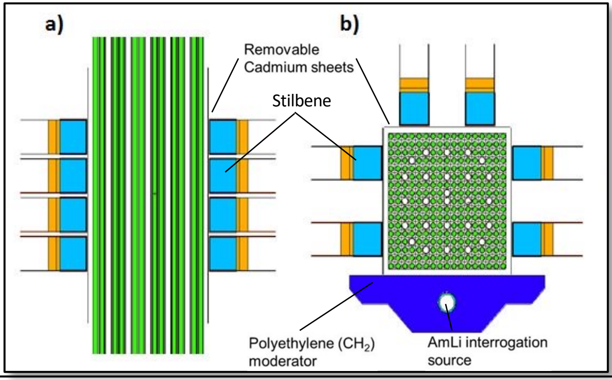
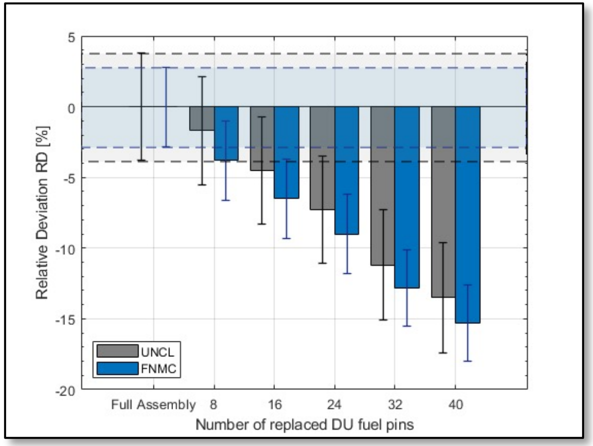


Image of the IAEA UNCL system (a), and the benchmarked MCNP simulation model (b)



Side-view (a) and top-view (b) of the FNMC system MCNP simulation model consisting of 30 2” Ø x 2” *trans*-stilbene detectors and an AmLi interrogation source

FNMC versus UNCL

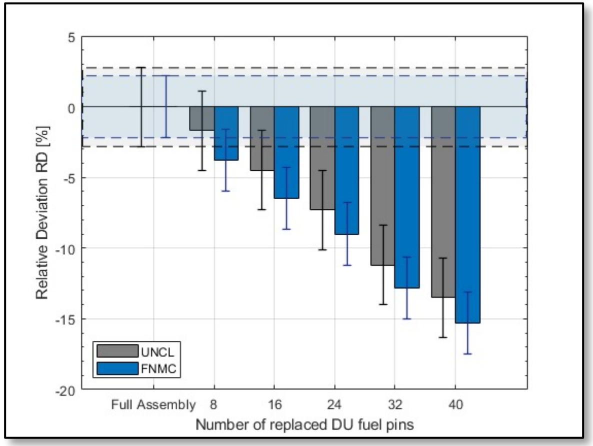


Simulated results for sensitivity of detecting diverted fuel pins for a 600-second assay time

System	Number of replaced LEU fuel pins				
UNCL	8	16	24	32	40
FNMC	8	16	24	32	40

FNMC system can **reduce assay time by one-third** due to improved measurement precision

→ Reduce operational cost
→ Improve sensitivity of detecting small-mass diversions
→ Increase number of assays



Simulated results for sensitivity of detecting diverted fuel pins for an 1800-second assay time

System	Number of replaced LEU fuel pins				
UNCL	8	16	24	32	40
FNMC	8	16	24	32	40

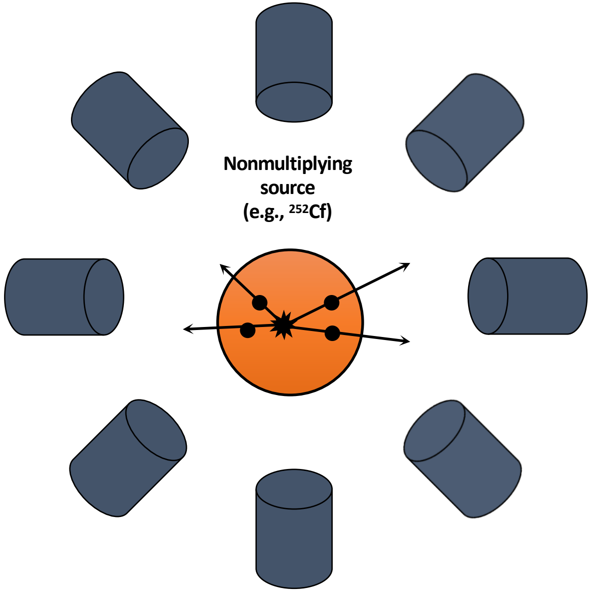
Nuclear Disarmament:

Can we ensure nuclear dismantlement while protecting sensitive information?

Neutron-neutron angular and energy-angle correlations: single-fission events

Well-known anisotropic distribution of fission neutrons from single spontaneous fission events

- More neutron-neutron coincidences observed at smaller (→ 0°) and larger (→ 180°), with a minimum at 90°
- Due to the kinematic boost received by the fission neutrons from the fully accelerated fission fragments
- Increased observed anisotropy for neutrons of higher energies



Neutron-neutron angular and energy-angle correlations: single-fission events

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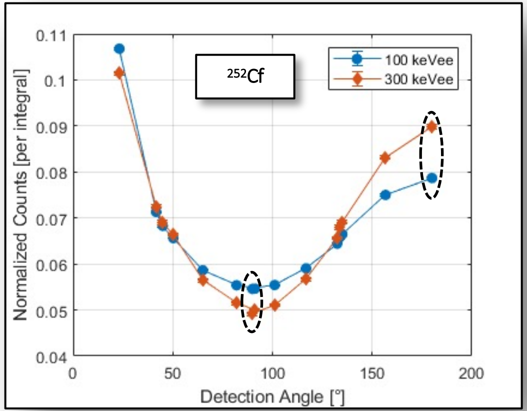
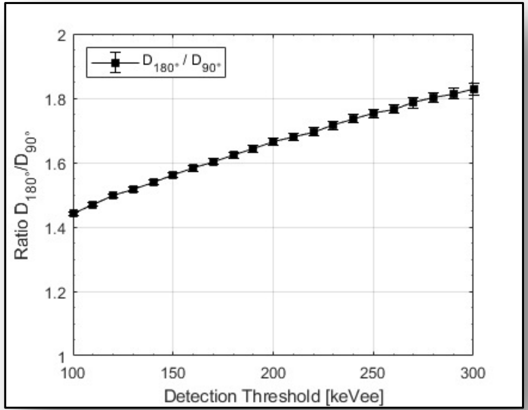


Measured neutron-neutron angular correlation from ^{252}Cf spontaneous fission neutrons for two different detection thresholds

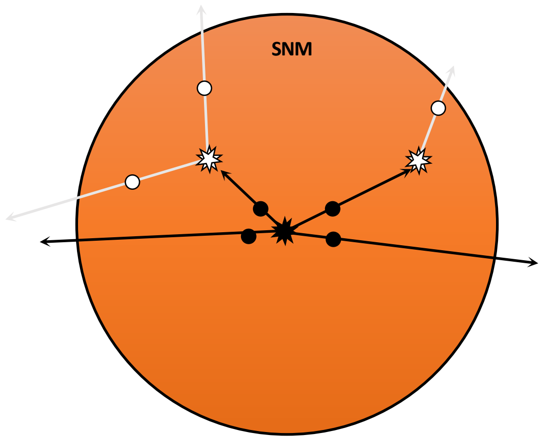


Measured neutron-neutron energy-angle correlation from ^{252}Cf spontaneous fission neutrons

Neutron-neutron anisotropy and fission chain length

Observed neutron-neutron angular and energy-angle correlations are expected to **diminish** as the fission chains **increase** in length

- Due to weakening of angular correlation between initiating neutron and the subsequently multiplied neutrons in the fission chain

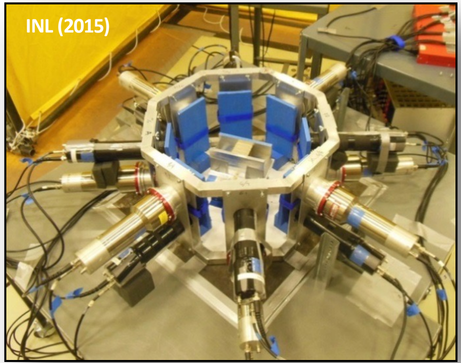


Expected to appear more isotropic as the multiplication increases

Passive NDA measurements of Pu-metal plates

120 total detector-pair combinations with nine unique angle groups (within ± 2°)

Applied detection threshold of 0.5, 1.0, and 1.5 MeV neutron-equivalent energy

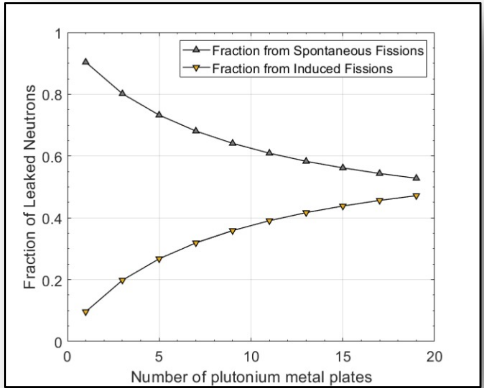


Passive measurements with 8 EJ-309 and 8 stilbene

Goal: characterize angular and energy-angle correlations as a function of leakage multiplication

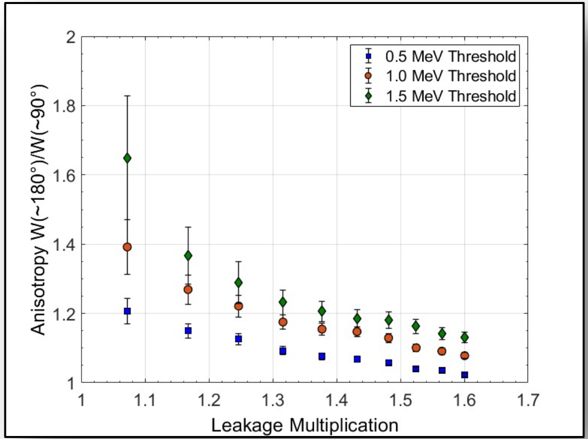
MCNP calculation of total and leakage multiplication

No. of PANN plates	Total multiplication	Leakage multiplication
1	1.1068(3)	1.0722(3)
3	1.2482(4)	1.1674(3)
5	1.3660(4)	1.2466(4)
7	1.4694(4)	1.3161(4)
9	1.5604(4)	1.3772(4)
11	1.6421(4)	1.4320(4)
13	1.7158(5)	1.4814(4)
15	1.7804(5)	1.5247(4)
17	1.8402(5)	1.5649(4)
19	1.8936(5)	1.6006(4)

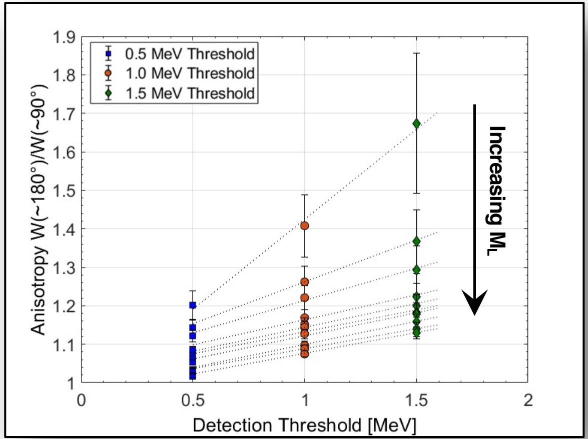


MCNP calculation showing fraction of leaked neutrons from spontaneous fission and induced fission events

Neutron-neutron angular and energy-angle correlations versus leakage multiplication



Measured neutron anisotropy as a function of the leakage multiplication at three different detection thresholds



Measured neutron anisotropy as a function of the detection threshold for all multiplying Pu-metal assemblies

Neutron-neutron coincidences become more **isotropic** as **multiplication increases**
Positive energy-angle correlation **diminishes** as **multiplication increases**

Can be used to characterize leakage multiplication for unknown SNM

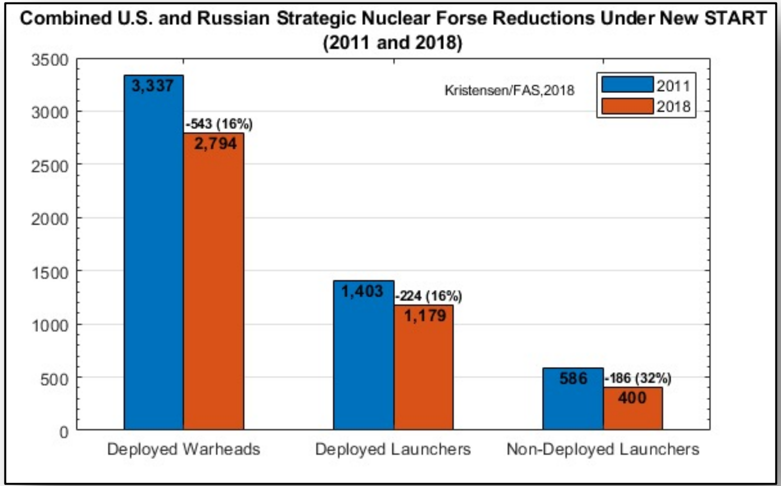
Verification of Current Nuclear Arms Control Treaties

Initiated in the late 1990s between Russia and the United States (START I, START II, SORT, etc.)

New Strategic Arms Reduction Treaty (New START): reduce number of deployed strategic nuclear weapons

Warheads are counted indirectly through the characteristic delivery vehicles they are associated with

Future disarmament treaties may limit the total number of nuclear weapons and warheads in the arsenal



May require inspection of **individual warheads and components**

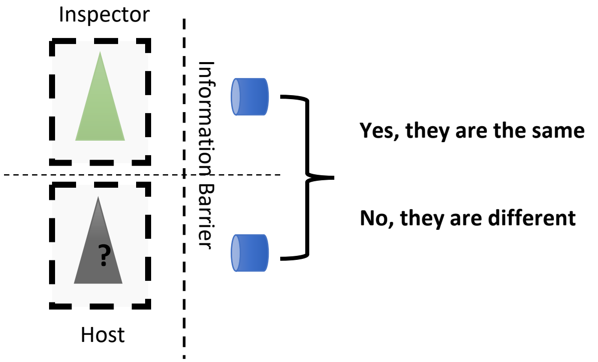
Verification Process: A Complex Transparency Problem

Nuclear weapon design is highly classified and must be protected for nonproliferation

→ **Confidently confirm authenticity of a warhead without revealing any sensitive information**

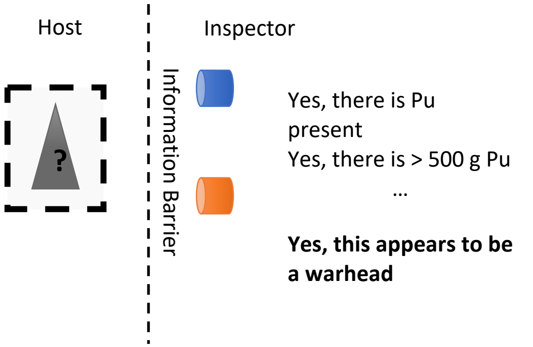
Template Approach
Acquire a unique "fingerprint" of the warhead under inspection, and compare it to a recorded template generated with an authentic ("golden copy") reference warhead

Confirm that the item under inspection is the *same* as the reference item



Attribute Approach
Measure intrinsic characteristics of nuclear weapons and their components

Confirm that the warhead under inspection appears to be an authentic warhead through observations of measured attributes



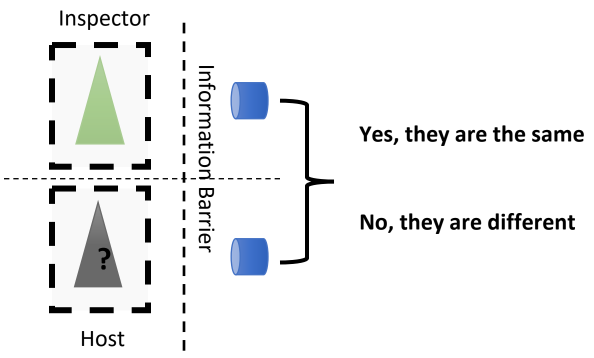
Template versus Attribute Approach

Template Approach
Considered more robust against “spoofs”

Beneficial for inspecting regime with numerous weapons of the same type

Requires storage of reference data

Requires agreed-upon reference item

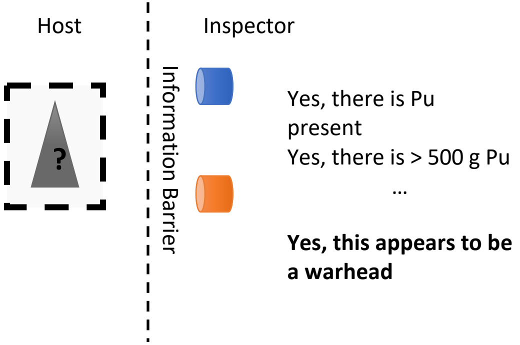


Attribute Approach
Does not require agreed-upon reference item

Does not require storage of reference data

Beneficial for inspecting regimes with similar but not the same type of weapons

Requires quantitative values and agreed-upon acceptance criteria



Attribute Approach

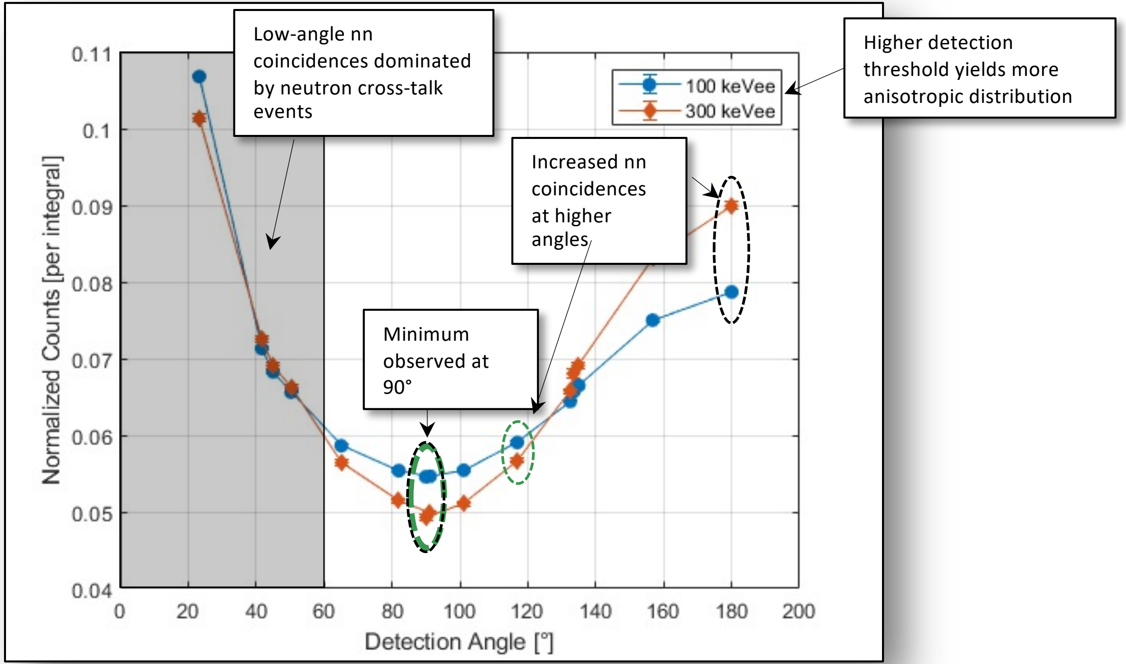
Ideal attribute for verification process

1. Easy to measure (simple system)
2. Difficult to spoof (robust)
3. Presence or absence of the attribute can be determined as a "yes" or "no" (information protection/barrier)
4. Indicative of fissile material (physically relevant to special nuclear material)

Attribute[1]	Measurement Type	Detector/System	Approach
Presence of Pu	Gamma-ray	HPGe	Spectrometry
Isotopics	Gamma-ray	HPGe	Spectrometry
Pu mass	Neutron	Neutron Multiplicity Counter	Point model estimation of fissile mass
Absence of oxide	Gamma-ray + Neutron	HPGe + Neutron Multiplicity Counter	Spectrometry + neutron singles rate
Age of Pu	Gamma-ray	HPGe	Spectrometry
Symmetry	Neutron	Neutron Multiplicity Counter	Count uniformity in all detectors
Presence of multiplying Pu	**Neutron**	**Fast-Neutron Multiplicity Counter**	**Prompt fission neutron anisotropy**

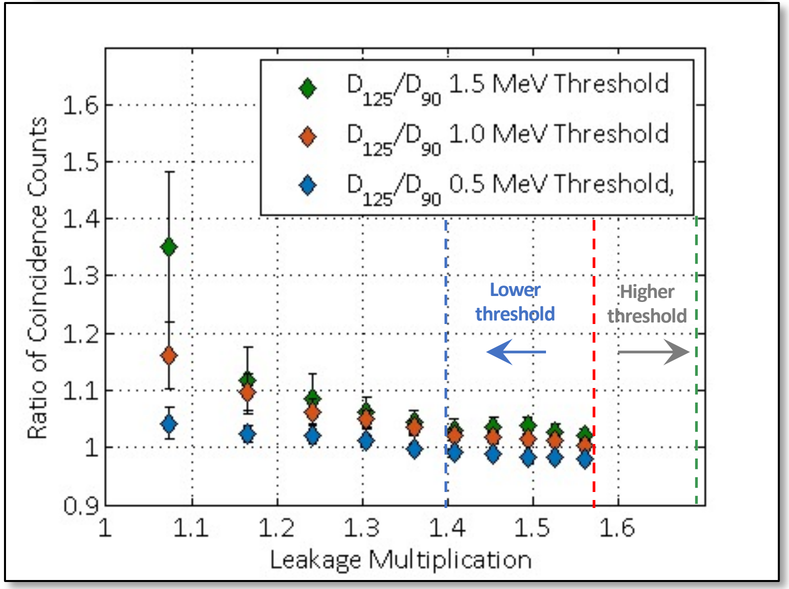
Can we detect the **presence of multiplying Pu** as a potential attribute for verification of future arms-control treaties?

A closer look at neutron-neutron angular distributions…



Neutron-neutron Anisotropy versus Energy Threshold

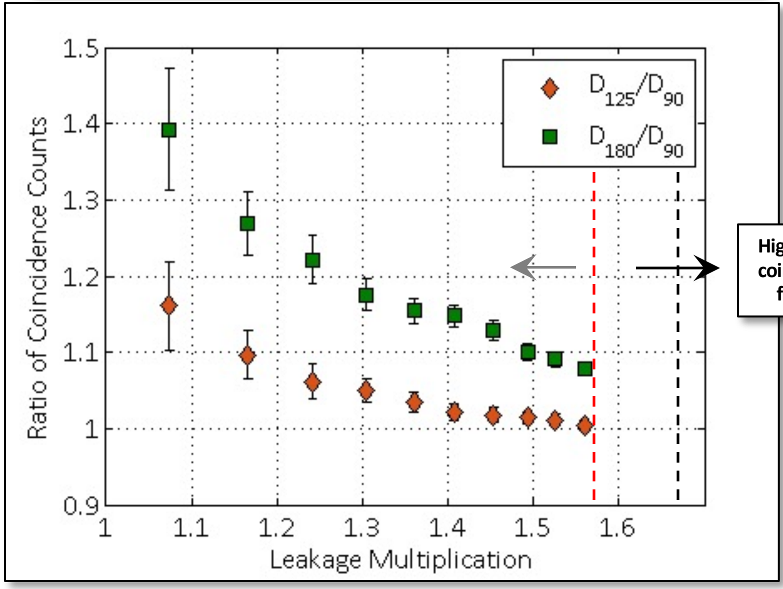
Prompt fission neutron anisotropy quantified using ratio of nn coincidences at 125° to those at 90° for various thresholds



Sensitivity of observed neutron anisotropy to multiplication dependent on detection thresholds
→ **"cutoff" multiplication shifts to the right (left) as detection threshold increases (decreases)**

Neutron-neutron Anisotropy versus Detection Angle

Prompt fission neutron anisotropy quantified using ratio of nn coincidences at 180° and 125° to those at 90° for 1 MeV threshold



Higher-angle coincidences for ratio

Sensitivity to multiplication increases when using higher angles for the ratio of nn coincidences
→ **"cutoff" multiplication shifts to the right when using higher-angle coincidences**

Conclusions

Nuclear safeguards and nonproliferation significant to ensure peaceful use of nuclear materials and associated technology

Technical methods must meet treaty requirements, and verifying future treaties must be technical plausible

Nuclear fission process is information rich; characterizing these signatures is an active area of research

Once characterized, can we develop techniques that leverage correlated signatures?

Transitioning technology to treaty-level applications can be difficult and slow, but there is a need for implementing novel approaches

Always be forward-thinking about adopting new technology to develop detectors, techniques, and theory for next-generation systems!